

## Critical Behavior of the Shear Resistance of Solid $^4\text{He}$ near a Structural Phase Transition

Inon Berent and Emil Polturak

*Physics Department, Technion-Israel Institute of Technology, Haifa 32000, Israel*

(Received 17 March 1998)

We describe experiments on plastic flow of solid  $^4\text{He}$  near the first order bcc-hcp transition. We find that the resistance to shear of the solid diminishes strongly near the transition. We are able to distinguish between plastic flow due to vacancies and due to dislocations, and show that the critical behavior of the solid near the transition is controlled by vacancy diffusion. The enhancement of the diffusion is consistent with a coupling of the vacancies to a phonon which softens near the transition. Possible implications of our findings on the understanding of melting are discussed. [S0031-9007(98)06732-5]

PACS numbers: 67.80.Mg, 62.20.Hg, 64.70.Dv

In an experiment aimed at measuring the plastic flow properties of solid He, we found that the shear resistance of the solid decreases dramatically as the temperature nears that of a bcc-hcp phase transition [1]. Over a temperature interval of 0.1 K, the plastic flow rate increased by  $1\frac{1}{2}$  orders of magnitude. In that experiment, the flow of solid took place by diffusion, yielding a value of the diffusion coefficient at the transition typical of a liquid. The loss of resistance to shear is the most prominent feature associated with melting. Theories of bulk melting proposed over the years rely on the sudden proliferation of crystalline defects in order to destabilize the lattice [2] so that the shear resistance vanishes. These include point defects (vacancies or interstitials) or line defects, namely, dislocations. However, this scenario was never confirmed experimentally, as the equilibrium density of defects in conventional solids is rather small, and it is impossible to approach the transition close enough to see critical behavior of the defects. The first order bcc-hcp structural phase transition in solid  $^4\text{He}$  offers several advantages in this type of study. First, it is weakly first order (the entropy change per particle is 12 times less than that associated with melting [3]), and consequently the temperature interval where pretransition effects can be observed is wider. One can therefore resolve how the shear resistance changes as the phase transition is approached. Second, the estimated density of vacancies [4] in  $^4\text{He}$  is significantly higher than what can be achieved in conventional solids even at the melting temperature. This condition of having a high defect density is another indispensable feature of the models of melting. Third, macroscopic single crystals of very high quality and purity can be grown. Furthermore, by investigating the plastic flow rate over a wide range of applied stresses, we can decide which type of defect, vacancies or dislocations, is responsible for the critical behavior near the transition (at low stress plastic flow results from vacancy diffusion, while at higher stress moving dislocations determine the rate). Thus, solid He is an excellent model system in which effects typical of the approach to the melting tran-

sition, such as loss of shear resistance and high defect density, can be conveniently studied. In this Letter we report the results of experiments on plastic flow carried out on solid  $^4\text{He}$  in equilibrium near bcc-hcp phase transition taking place at  $T = 1.772$  K on the melting curve.

The measuring technique detects the change of the position of a wire, submerged in the crystal, driven by a constant force. Plastic properties of solid He were investigated, by using motion of a solid body, by several groups in the past [5,6]; however, ours is the first such experiment near a phase transition. In most of this work we used a superconducting wire of diameter  $80\ \mu\text{m}$  as the moving solid body (in some of the earlier experiments we used a  $24\ \mu\text{m}$  wire [7]). The experimental cell was situated in a constant uniform magnetic field. A dc current was driven through the wire, so that it moved under the Lorentz force. This arrangement is drawn schematically in Fig. 1. To prevent any Ohmic heating, the range of currents used was less than 10% of the wire critical current. The moving part of the wire was in a shape of a semicircle of 28 mm radius. To detect the wire position, a low amplitude audio frequency ac current was superimposed on the dc current. A 600 turn receiver coil was situated in parallel to the plane of moving wire, 1 mm apart, concentric with the wire curvature. Changes in the wire position were detected by measuring the changes in the induced voltage on the receiver coil. The lowest wire velocities measured were in the  $10^{-9}$  cm/sec range, while near  $T_c$  the phase transition temperature and under

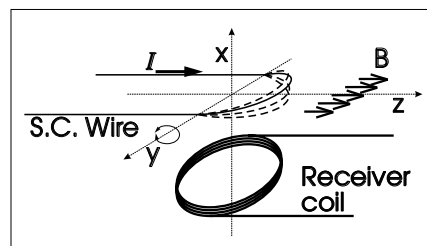


FIG. 1. Schematic drawing of the moving wire assembly.

high stress the wire velocity exceeded  $10^{-5}$  cm/sec. In addition to the wire assembly, the cell contained a parallel plate capacitor which was used to measure the level of the solid in the cell and its density, a Straty-Adams capacitive pressure gauge, and a cold valve which isolated the cell from the filling line during measurements. High quality crystals were grown at a constant temperature, at a rate which varied between  $1.6 \times 10^{-4}$  cm/sec and  $4 \times 10^{-5}$  cm/sec. Growth was usually terminated after the solid filled the cell to a level 4 mm above the wire. After the end of growth, the crystal was allowed to anneal for about 12 h before measurement began. In total, we grew about 70 crystals, out of which half were of good enough quality to be measured (good crystals showed no measurable yield stress, indicating that the initial density of dislocation was very small). A measurement was initiated by applying a dc current to the wire while holding the temperature constant. Under small stresses, a steady motion was observed, from which the steady state velocity of the wire was determined. At higher stresses, where the motion is determined by dislocation motion, a short transient was seen, resulting from an initial rearrangement of the dislocations near the wire (Andrade creep). After less than 2 min, the wire settled into a steady motion which was followed for another 30 min in order to determine the steady state velocity. The stress exerted by the wire under the range of forces used ranged from  $1.8 \times 10^2$  to  $1.5 \times 10^4$  dyn/cm<sup>2</sup>. Results obtained over time using different crystals were reproducible within the scatter of the data.

Figure 2 shows a typical dependence of the measured velocity on the applied stress in the two solid phases. As the inset shows, under low stresses (smaller than  $6 \times 10^3$  dyn/cm<sup>2</sup>) the velocity of the wire is linearly proportional to the applied stress, while at higher stress the velocity becomes proportional to a higher power of  $\sigma$ .

The linear dependence of the velocity  $v$  on the applied stress  $\sigma$  seen at low stresses is characteristic of the Nabbaro-Herring [8,9] mechanism, where the mass flow takes place through counterdiffusion of vacancies and atoms due to a vacancy concentration gradient. In this picture, the velocity of the wire  $v$  depends linearly on the applied stress  $\sigma$  [5,7]:

$$\frac{v}{\Omega} = D_s \frac{\sigma}{k_B T l}. \quad (1)$$

Here,  $\Omega$  is the atomic volume,  $D_s$  is the self-diffusion coefficient,  $T$  is the temperature, and  $l$  is the wire diameter. In this picture, atomic diffusion takes place between the front of the wire where the stress is high and the back of the wire where the stress is low. We demonstrate the validity of this picture by plotting in Fig. 3 all the data obtained in the linear regime using different diameter wires and different stresses, scaled according to Eq. (1). Figure 3 also shows the temperature dependence of the wire velocity for high ( $\sigma = 1.5 \times$

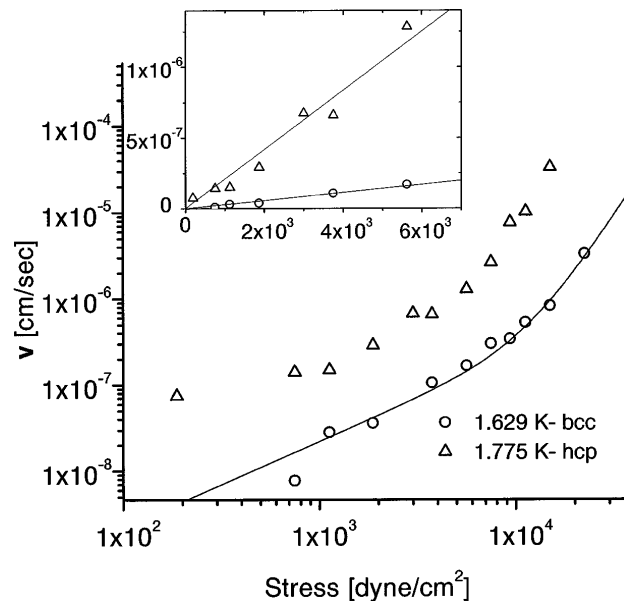


FIG. 2. The velocity of the wire as a function of the applied stress at two temperatures. Solid line is a fit to the data involving both vacancies and dislocations (see text). The inset contains a linear-linear plot of the small stress data, showing that in this regime the velocity is linearly proportional to the applied stress.

$10^4$  dyn/cm<sup>2</sup>) applied stress. The temperature at which the peak in  $v$  in Fig. 3 occurs coincides with the published coordinates of the hcp-bcc transition to better than 1 mK. This identification is important in order to rule out the possibility that the behavior shown here has anything to do with the liquid phase. First, we discuss  $D_s$ , which was calculated from the data obtained in the linear regime using Eq. (1). Writing  $D_s = D_0 \exp[-E(T)/kT]$ , and assuming that the prefactor  $D_0$  does not change with temperature, we can translate the increase of  $D_s$  near the transition to a reduction of  $E(T)$ , the activation energy for self-diffusion. (In the band model of vacancy diffusion [4] in solid He,  $D_0 \propto T^3$ , which leads to a 20% change of  $D_s$  in the region of interest between 1.7 and 1.8 K. The velocity of the wire, which is proportional to  $D_s$ , changes by a factor of 15 in this range, so we can indeed neglect the temperature dependence of  $D_0$ .) From fits to the data, shown as the solid lines in Fig. 3, we found that close to the transition in the bcc phase  $E(T)$  decreases linearly with  $t \equiv (T_c - T)/T_c$  (see inset of Fig. 3). Away from the transition,  $E(T)/k_B = 14.8$  K, in agreement with previous work [5], while near the transition it decreases to a value of 11.8 K. The behavior is qualitatively similar in the hcp phase, with  $E(T)$  increasing by 6 K between  $T_c$  and temperatures well away from the transition. In this paper, we shall discuss only the bcc phase.

Schober *et al.* [10] have shown that  $E(T)$  can be thought of as the elastic energy needed to deform the lattice so that an atom can diffuse into a nearest neighbor vacant site. This elastic energy is expressed in terms

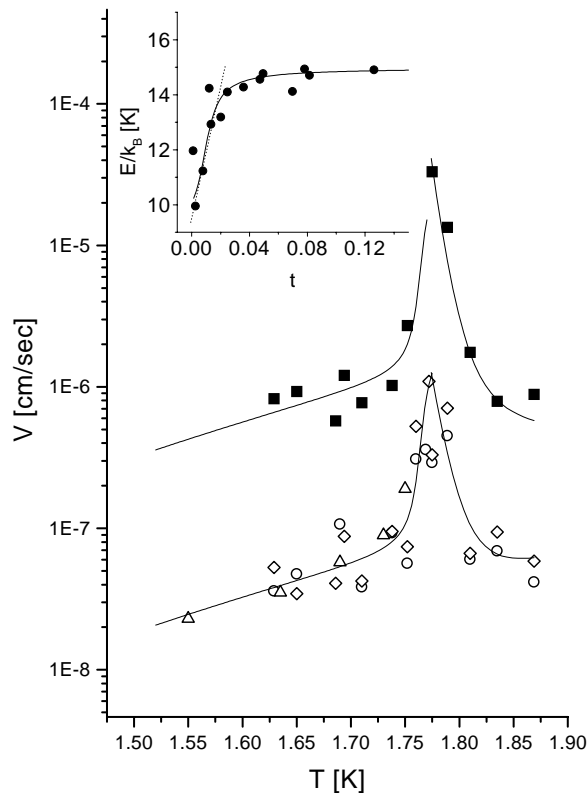


FIG. 3. Wire velocity  $v$  as a function of the temperature. Open symbols include data in the linear response regime, obtained using two different wires with different diameters. Closed symbols are typical data obtained in the nonlinear regime (large stress). The solid lines are fits to the data using the phonon assisted diffusion concept. The inset shows activation energy for self-diffusion near  $T_c$ , plotted against reduced temperature in the bcc phase. The dashed line shows the linear behavior, while the solid line is an empirical fit to the data.

of the phonon spectrum, where  $E(T)$  depends on the frequency of any particular phonon as  $\omega^2$ . The observed decrease of  $E(T)$  implies that the phonons soften near  $T_c$ . A linear decrease of  $E(T)$  near  $T_c$  is predicted by the Landau theory [11] of a structural phase transition driven by a *single* phonon soft mode. It turns out that softening of specific phonons in the bcc phase is a common feature in many metals exhibiting enhanced diffusion near a bcc-hcp transformation. It proved possible to pinpoint the mechanism of diffusion in these metals using quasielastic neutron scattering [12]. These experiments show that self-diffusion is a phonon assisted nearest neighbor atom-vacancy exchange. Petry *et al.* [12,13] found that there is one dominant phonon assisting the diffusion, the  $1/2$  (1, 1, 0) transverse phonon, and that it also softens appreciably as the phase transition temperature is approached. The measured phonon spectrum of bcc  $^4\text{He}$  is very similar to that of metals. The measured spectrum [14,15] is in very good agreement with theoretical calculations [16] done at  $T = 0$ , except for the

same  $(\xi, \xi, 0)$  transverse phonon branch, which appears to be much softer. We therefore take the view that the mechanism of self-diffusion in bcc  $^4\text{He}$  is the same as in bcc metals, and the decrease of  $E(T)$  near the transition results from the softening of this phonon. It is important to stress that in metals, where formation energy of a vacancy, typically  $0.5-1 \text{ eV} \gg \hbar\omega$ , which is  $\sim 10 \text{ meV}$ , the phonon assisted diffusion model refers to changes of the energy associated with the migration of vacancies, without changing their density. In solid He, the formation energy ( $\approx 1 \text{ meV}$  [17]) is quite close to  $\hbar\omega$  ( $\approx 0.5 \text{ meV}$ ), so that both the density and the mobility of vacancies could be affected.

We now turn to the stress dependence of the velocity at larger stresses. We find that  $v \propto \sigma^q$  in this region, with  $q = 3.5 \pm 0.3$ . The uncertainty in the value of  $q$  comes from fitting data sets obtained at different temperatures. The solid line in Fig. 2 shows a fit to a form  $V(\sigma) = a\sigma + b\sigma^{3.5}$ , where  $a$  is determined from Eq. (1) and  $b$  is a parameter. Such dependence is characteristic of combined motion due to self-diffusion and dislocation climb. In polycrystalline bcc  $^3\text{He}$ , the strain rate in the range of stresses and velocities controlled by climb of dislocations [18] was found to be proportional to  $\sigma^{3.8}$ , and in polycrystalline hcp  $^4\text{He}$  to  $\sigma^4$  [6]. Models describing dislocation climb [8,19] give power law dependence in this range, and there could be more than one mechanism that controls the flow in the same crystal. The creep rate of isolated dislocations, determined by the rate at which the dislocation climbs, is linearly proportional to the stress, but, if a dislocation network exists, the creep rate is proportional to higher power of the stress. In both cases, climb of dislocations always occurs through emission or absorption of point defects, and is therefore ultimately controlled by self-diffusion. We therefore expect the velocity of the wire to be proportional to the diffusion coefficient not only at low stresses, but through the whole range of measured velocities. In order to see whether the increase of the velocity in the transition region reflects only the enhanced diffusion, or if there is another mechanism related only to dislocations, we assumed that the velocity can be written in the form  $v(T, \sigma) = f(T)g(\sigma)$ . If the anomalous dependence near  $T_c$  is only due to vacancies, then dividing the data by  $f(T) = D_s/T$  should effectively remove the temperature dependence from the data. In Fig. 4 we show the data before and after performing this operation. It seems indeed that the operation effectively removes any temperature dependence. The variable that is traditionally used to describe dislocations under applied stress is the ratio  $\sigma/\mu$ , where  $\mu$  is the shear modulus of the material. One of the earliest models for melting [20] associated melting with vanishing of  $\mu$  at the transition temperature. In many solids, including helium,  $\mu$  decreases as the melting temperature is approached to about half of its low temperature value. We thus checked whether the scatter in  $g(\sigma)$  can be reduced further by inserting an indirect dependence on

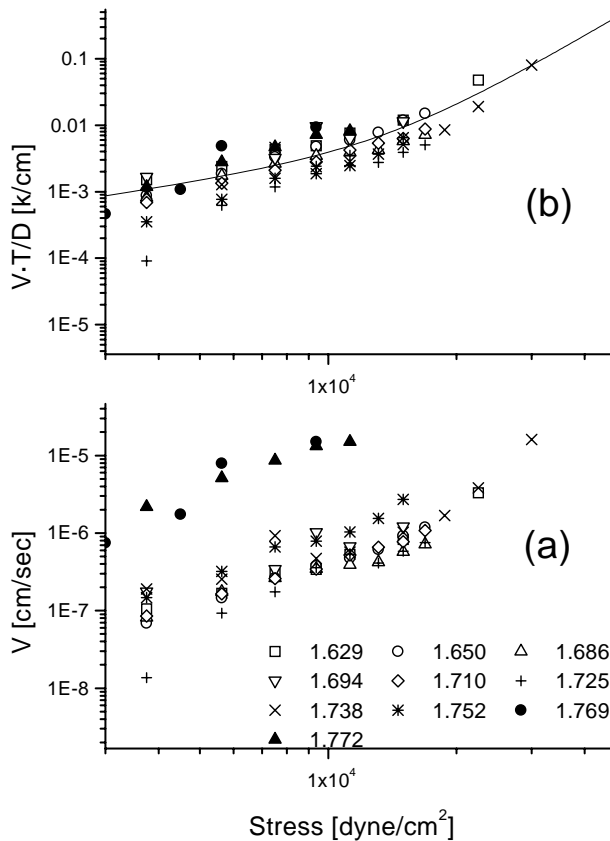


FIG. 4. Velocity of the wire vs  $\sigma$  in the nonlinear regime. (a) Raw data at various temperatures. (b) Same data scaled by  $D/T$ . Solid line in (b) is a fit to the combined dependence of  $v$  on self-diffusion and dislocation climb.

the temperature, by plotting the data vs  $(\sigma/\mu)$ , and then letting  $\mu$  change proportionally to  $E(T)$ , or alternately to some power of the reduced temperature. We found no significant change in the scatter of  $g(\sigma)$  in all these trials. We conclude therefore that the enhanced plastic flow near  $T_c$  is fundamentally due to vacancies, which influence the dislocations by controlling their climb rate.

In conclusion, it seems that the primary decrease in resistance to shear can be attributed only to vacancies, and the dislocation population is not fundamentally associated with the phase transition. Second, the critical behavior of plastic flow near  $T_c$  can be explained by the same model relating the bcc-hcp phase transition in metals to a softening of a phonon with temperature. Regarding the implications of this experiment to the understanding of melting, it appears that it is sufficient for one phonon to soften in order to destabilize the lattice so that it loses its resistance to shear. This is a refinement of the view expressed in the Lindemann theory, which treats all the

phonons on an equal footing. Furthermore, the relevant phonons are those near the edge of the Brillouin zone, and not an averaged spectrum as expressed in the Debye picture. The close interrelation between phonons and point defects indicates that one should incorporate both into an improved model of melting.

We are indebted to M. Ayalon, S. Hoida, and M. Kaganovitch for their assistance. This research was supported by the Israel Science Foundation, and by the Technion VP Fund for the Promotion of Research.

- [1] E. Polturak, I. Schuster, I. Berent, Y. Carmi, S. G. Lipson, and B. Chabaud, *J. Low Temp. Phys.* **101**, 177 (1995).
- [2] R. M. Cotterill, E. J. Jensen, and W. D. Kristensen, in *Anharmonic Lattices, Structural Transitions and Melting*, edited by T. Riste, NATO Advanced Study Institutes, Ser. E (Noordhoff, Leiden, 1974).
- [3] J. K. Hoffer, W. R. Gardner, C. G. Waterfield, and N. E. Philips, *J. Low Temp. Phys.* **23**, 62 (1976).
- [4] C. A. Burns and J. M. Goodkind, *J. Low Temp. Phys.* **95**, 695 (1994).
- [5] N. E. Dyumin, N. V. Zuev, and S. V. Svatko, and V. N. Grigor'ev, *Low Temp. Phys.* **17**, 468 (1991).
- [6] H. Suzuki, *J. Phys. Soc. Jpn.* **42**, 1865 (1977).
- [7] I. Berent and E. Polturak, *J. Low Temp. Phys.* (to be published).
- [8] F. R. N. Nabarro, *Theory of Crystal Dislocation* (Oxford University Press, New York, London, 1967).
- [9] C. Herring, *J. Appl. Phys.* **21**, 437 (1950).
- [10] H. R. Schober, W. Petry, and J. Trampenau, *J. Phys. Condens. Matter* **4**, 9321 (1992).
- [11] R. A. Cowley, *Adv. Phys.* **29**, 1 (1980).
- [12] W. Petry, A. Heiming, J. Tampeanu, and G. Vogl, *Defects and Diffusion Forum*, edited by F. J. Kedevs and D. L. Beke (Sci-Tech Publ., Brookfield, VT, 1989), Vols. 66–69, p. 157.
- [13] W. Petry, A. Heiming, J. Tampeanu, M. Alba, C. Herzig, H. R. Schober, and G. Vogl, *Phys. Rev. B* **43**, 10933 (1991).
- [14] V. N. Minkiewicz, T. A. Kitchens, G. B. Shirane, and E. B. Osgood, *Phys. Rev. A* **8**, 1513 (1973).
- [15] E. B. Osgood, V. N. Minkiewicz, T. A. Kitchens, and G. Shirane, *Phys. Rev. A* **5**, 1537 (1972).
- [16] H. R. Glyde, *J. Low Temp. Phys.* **3**, 559 (1970).
- [17] P. R. Granfors, B. A. Fraass, and R. O. Simmons, *J. Low Temp. Phys.* **67**, 353 (1987).
- [18] A. Sakai, Y. Nishioka, and H. Suzuki, *J. Phys. Soc. Jpn.* **46**, 881 (1979).
- [19] J. Weertman and J. R. Weertman, *Physical Metallurgy*, edited by R. W. Cahn (North-Holland Publishing, Amsterdam, 1965).
- [20] M. J. Born, *Chem. Phys.* **7**, 591 (1939).